

Topological phase transition induced by random substitution

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The transition from topologically nontrivial to a trivial state is studied by first-principles calculations on bulk zinc-blende type $(\text{Hg}_{1-x}\text{Zn}_x)(\text{Te}_{1-x}\text{S}_x)$ disordered alloy series. The random chemical disorder was treated by means of the Coherent Potential Approximation. We found that although the phase transition occurs at the strongest disorder regime ($x \approx 0.5$), it is still manifested by well-defined Bloch states forming a clear Dirac cone at the Fermi energy of the bulk disordered material. The computed residual resistivity tensor confirm the topologically-nontrivial state of the HgTe-rich ($x < 0.5$), and the trivial state of the ZnS-rich alloy series ($x > 0.5$) by exhibiting the quantized behavior of the off-diagonal spin-projected component, independently on the concentration x .

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I. INTRODUCTION

The most attractive property of topologically-nontrivial insulating materials is the presence of the electronic chiral states at their surface, forming the so-called Dirac cone, which gives rise to the quantized non-dissipative and fully spin-polarized surface current [1]. This surface property is exploited in band structure calculations, which intend to prove the trivial or nontrivial topology of a given system [2]. Since the topology is directly related to the bulk band structure, those approaches which can predict the topological class of a given material based on purely bulk information are more practical as being computationally less demanding compared to realistic surface calculations. Thus, in the search for novel materials such theoretical approaches where the topological phase transition can be straightforwardly investigated in the bulk are very useful tools, that are able to provide supporting data to motivate experimental investigations on unexplored new compounds.

Indeed, from the point of view of the band structure, the manifestation of the trivial-nontrivial phase transition is always marked by the existence of a Dirac point, independently on the nature of the order parameter responsible for the assignment of a given material to a certain topological class. In contrast to expensive and tedious experimental methods, in theoretical studies these parameters can be easily tuned. For example, the trivial-nontrivial phase transition can be simply induced by changing the amplitude of the spin-orbit coupling, or the crystal-field splitting (e.g. through the change of the lattice constant) [3]. In case of well-defined Bloch states one can also make use of the parity analysis of the eigenstates [4]. However, the most problematic

cases are those, where the local translational symmetry is broken due to random fluctuations of certain degrees of freedom, which are inevitably always present in real materials. These fluctuations can be caused by random chemical disorder, various dynamical fluctuations as e.g. phonons or magnons, or strong local electron correlations which are specific for systems with strong spin-orbit coupling. All these mechanisms lead to a localization of the electronic states which can no longer be expressed in the form of Bloch waves. This makes the straightforward analysis of the parity of the eigenstates inapplicable. Also, in the ordered class of materials so far only a few stable non-trivial compounds have been found. Therefore, the search for topological insulators among disordered systems will gain an increased importance. This will be especially true for materials with application relevance, since disordered systems are easier to be mass produced, and despite structural disorder their topological edge states are robustly protected against any time-reversal symmetric perturbation. Hence, by extending our field of interest towards disordered systems many new topologically-nontrivial materials can be found.

The practical tools for the first-principles studies on the disordered systems are typically provided by the effective mean-field theories, such as e.g. the Dynamical Mean-Field Theory (DMFT) [5] or via the Coherent Potential Approximation (CPA) [6, 7]. The former takes into account the local dynamical electronic correlations, and the latter efficiently describes the random chemical disorder. Each implementation of the mean-field theory incorporates the effect of a certain type of random fluctuations in the form of an energy-dependent complex-valued local potential (the so-called self-energy) added to the real-valued lattice-symmetric Kohn-Sham potential. This leads to an energy-dependent shift and to a broadening of the electronic states, seen in the band structures calculated with DMFT or CPA.

In the following we would like to demonstrate the

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power of the CPA as a practical adiabatic technique, which allows to determine the topological class of a given compound by its gradual transformation into another system, for which the topological class is already well established. In contrast to complicated surface calculations such bulk simulations can be easily performed within the bulk regime, thus being rather simple and reliable. In addition, by observing the topological phase transition in the bulk provides a wide range of new information regarding the scenario occurring on the borderline between non-trivial and trivial systems, where the mixing and interdiffusion often takes place in the quantum well structure formed by these two materials [8, 9].

In the following example we will consider the sequence of random alloys between the prototypical non-trivial gapless semiconductor HgTe and the trivial ZnS insulator, which are both non-magnetic binaries of the zinc-blende type. Since Zn is isovalent to Hg and S to Te, the intermediate alloys $(\text{Hg}_{1-x}\text{Zn}_x)(\text{Te}_{1-x}\text{S}_x)$ must be non-magnetic semiconductors, thus keeping the time-reversal symmetry of the total wave function. Although here we are investigating the aforementioned alloy series, we have to point out that all features of the topological phase transition shown in the current work can be transferred with no restriction to other systems, like for e.g. the rich family of the ternary Heusler semiconductors where many topologically non-trivial compounds were already identified [3].

II. COMPUTATIONAL METHODOLOGY

Until now the CPA (and its extensions) remains the only widely applicable alloy technique which incorporates the effects of the energy-dependent shift and lifetime broadening. Both are essential features of the electron localization caused by chemical disorder, which are not accessible via other theories, as e.g. the VCA (virtual crystal approximation) [10] or supercell calculations. Although the CPA method has lots of advantages, it has a typical shortcoming, namely in its original “single-site” formulation the effect of the local environment is absent. This bottleneck of CPA can be remediated, however, by employing its non-local extensions [11–13]. Regardless, we would like to emphasize that for the case of isovalent substitution the single-site CPA remains a quite good approximation even in the diluted limit, since the isovalent atoms intermix without additional environmental preference as long as the zinc-blende type crystal structure is preserved.

All electronic structure and spin-Hall transport coefficient calculations presented below were carried out using the fully-relativistic Korringa-Kohn-Rostoker (KKR) Green’s function method implemented within the SPR-KKR package [14] employing the density functional theory framework. The exchange and correlation was treated using the Vosko-Wilk-Nusair form of the local density approximation (LDA) [15]. LDA is known for

its typical underestimation of the band gaps of semiconductors. However, in case of our study this issue is not critical, since the observed trends are qualitatively well described even with LDA. The lattice constants for the alloys were derived by linear interpolation between the experimental lattice parameters of the pure HgTe and ZnS compounds as a function of the concentration of the constituents. Due to the multiple-scattering construction of the Green’s function [16], the method provides a suitable base for mean-field approaches like CPA. Since the electronic structure of disordered materials in general does not show well defined Bloch eigenstates, their electronic structure is described by the Bloch-spectral function (BSF) defined by the imaginary part of the alloy Green’s function that is diagonal in momentum space [16].

III. RESULTS AND DISCUSSION

In order to study the topological phase transition in bulk 3D materials, we have performed band structure calculations and we have investigated the spin-resolved transport properties of several quaternary alloys in the series of $(\text{Hg}_{1-x}\text{Zn}_x)(\text{Te}_{1-x}\text{S}_x)$. First, we present the analysis of the BSF computed for the most relevant alloy compositions, which are shown in Fig. 1. Since the band structure of pure HgTe ($x = 0$) was established long ago [17], here we just briefly mention the most characteristic features in the vicinity of the Γ point: the Te 5*p*-band is split due to spin-orbit coupling (SOC) into a Γ_8 state sitting right at E_F and a Γ_7 state, which is located about 1.2 eV below Γ_8 . The Hg 6*s*-band corresponding to Γ_6 is placed by the crystal field (CF) between the two aforementioned states, at about -0.8 eV from E_F . This negative value, often named “band inversion”, drives HgTe into a non-trivial class, resulting from the combined effect of the strong SOC and small CF. For $x > 0$ the symmetry analysis cannot be applied directly, since there are no pure Bloch eigenstates, i.e. in the disordered regime the delta-like poles of the BSF turn into overlapping Lorentzians spread over the whole (\vec{k}, E) -space. However, it is still interesting to trace the evolution of their maxima in analogy to the dispersion relation $E(\vec{k})$ of the pure material. By calculations we will show that the SOC and CF can be tuned as a function of composition [3]. Upon gradual substitution of Hg with Zn, and Te with S we effectively decrease the SOC and increase the CF. Hence, both Γ_7 - and Γ_6 -like states rise in energy (see Fig. 1). In the ZnS-poor regime the decrease of SOC acts very efficiently on the BSF, and at about $x = 0.3$ the Γ_7 -like state touches the Γ_6 -like state at roughly -0.5 eV below E_F . As far as there is no change of occupancies this does not affect the topological class of the system. By further substitution the Γ_6 state continues to rise towards E_F , being strongly influenced by the CF, whereas the effective decrease of SOC does not affect the Γ_7 -like state considerably. As shown by the

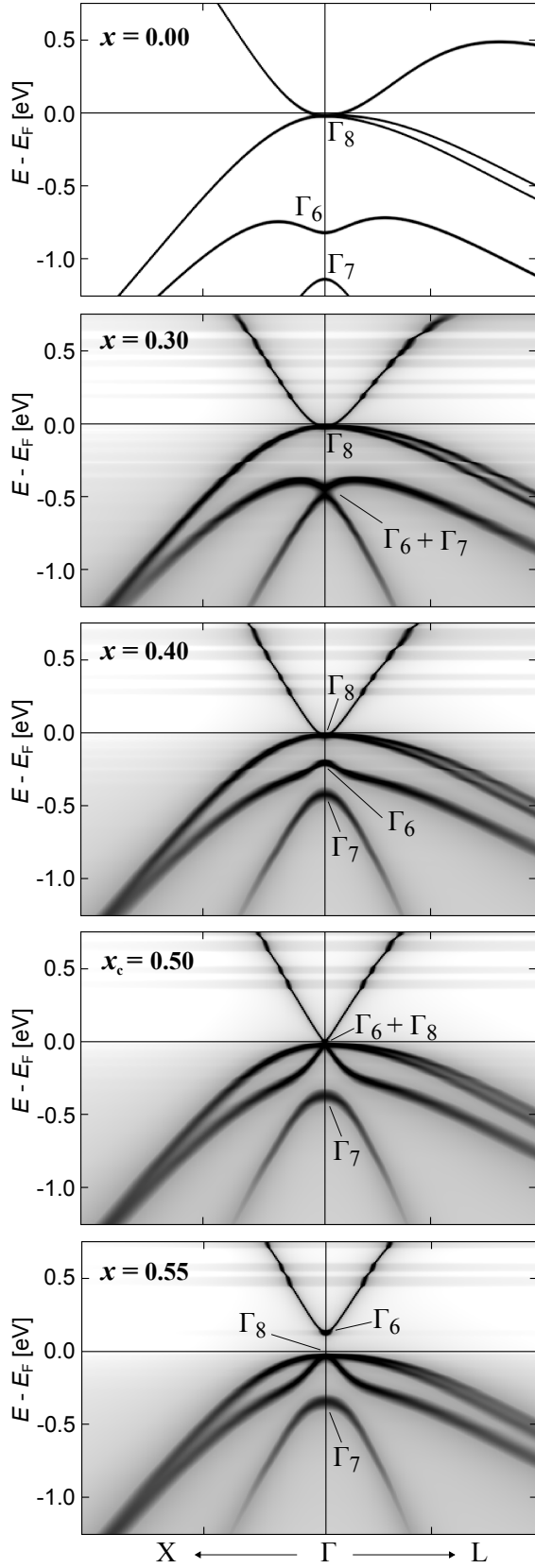


FIG. 1. Evolution of the Bloch-spectral function (along $X-\Gamma-L$) in the alloy series $(\text{Hg}_{1-x}\text{Zn}_x)(\text{Te}_{1-x}\text{S}_x)$. The critical composition $x_c = 0.5$ corresponds to the topological phase transition marked by a Dirac cone.

snapshot at the intermediate concentration $x = 0.4$, the band alignment in this regime is equivalent to the original pure HgTe case.

At the “critical” concentration of the random substitution $x_c = 0.5$ the system undergoes a topological phase transition manifested by the Dirac cone formed by a mixture of Γ_8 - and Γ_6 -like states. Obviously the edge states with linear dispersion at the surface of a topological insulator and the states forming the Dirac cone in the disordered bulk manifest the same transition mechanism. The only qualitative difference is that due to the bulk translational symmetry, we observe two replica of Dirac cones with opposite spins superimposed, whereas due to the break of space-reversal symmetry at the surface only a single cone remains, which exposes the adiabatic spin-current.

In the ZnS-rich regime ($x > 0.5$) which is topologically trivial due to the emptying of Γ_6 -symmetric state, the evolution of the energy levels are basically unaffected by x : Γ_7 is pinned to Γ_8 at E_F due to vanishing SOC, and the unoccupied Γ_6 state continues to move up increasing the band gap width. It is easy to see that in this way we will arrive to the band structure of pure ZnS, which is rather well known [18, 19]. Within this regime the electronic structure is close to the topologically “intermediate” pure compounds similar to CdTe [17] or CdSe [20], which are close to the borderline of trivial and topological insulators.

The energy distribution of the lifetime broadening can be understood by noticing that it scales with the band curvature $\sim \partial^2 E / \partial k^2$, i.e. with the effective mass. Indeed, the corresponding shades represent a weighted superposition of parabolic-like heavy bands exhibited by both pure HgTe and ZnS. In contrast to this, the appearance of Bloch-like states with linear dispersion, –i.e. the massless states– are essentially a new ordering feature arising from the quantum interference. This situation is very evident for the critical composition $x_c = 0.5$ which corresponds to the appearance of the conical dispersion and simultaneously most strongly influenced by disorder.

To verify that this is indeed a topological phase transition, in the following we will analyze the tensor of residual conductivity calculated by means of the general Kubo and Kubo-Středa formalism [7, 21]. In order to access the spin-resolved components $\sigma_{xy}^{\uparrow(\downarrow)}$ of the transversal conductivity, where the spin polarization $\uparrow(\downarrow)$ refers to the z -axis, we employed a relativistic scheme suggested recently [21, 22]. As it follows from Fig. 2 (a), the diagonal component σ_{xx} scales almost as $\sim x^{-1}$ from infinity (in the pure limit HgTe is a metal in our calculations) down to the values, which within a strong disorder regime become already comparable with the spin-Hall component σ_{xy}^{\uparrow} . On the ZnS-rich side ($x > 0.5$) both components quickly drop down almost to zero as a function of x , since the band gap starts to open. Due to the delocalization which takes place both in the \vec{k} - as well as in the energy space, this drop is not abrupt. Moreover, we notice that σ_{xy}^{\uparrow} does change its sign before it vanishes, which can be

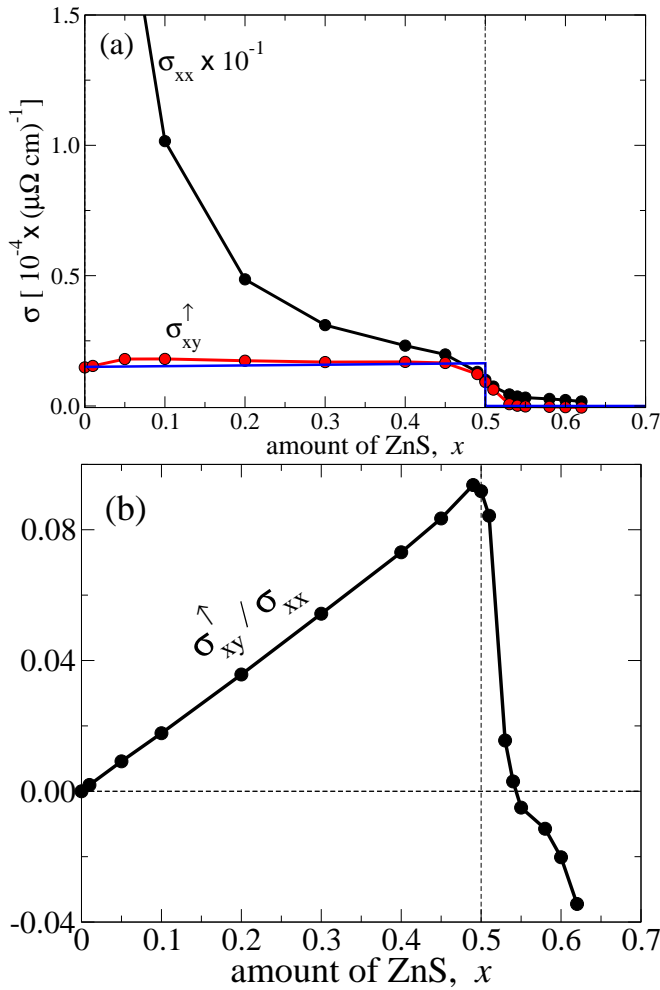


FIG. 2. (color online) (a) Diagonal (σ_{xx} , black) and spin-projected off-diagonal (σ_{xy}^\uparrow , red) components of the conductivity tensor calculated as a function of x in the disordered $(\text{Hg}_{1-x}\text{Zn}_x)(\text{Te}_{1-x}\text{S}_x)$ alloy series. These results are compared to the quantum of the 3D spin-Hall conductivity marked by the blue line. (b) The spin-Hall ratio $\sigma_{xy}^\uparrow / \sigma_{xx}$ plotted as a function of composition x .

seen from the dependency of spin-Hall ratios (shown in Fig. 2 (b)). The sign change is connected to the reversal of the Hall chirality after the cone center crosses the Fermi energy E_F .

In contrast to the diagonal component, the spin-Hall conductivity stays almost independent from the composition until the topological phase transition takes place, which indicates the quantized origin of σ_{xy}^\uparrow . Indeed, since σ_{xy}^\uparrow is calculated within the Kubo-formalism, together with the intrinsic quantized contribution (connected with the Berry curvature), it contains also extrinsic contributions (attributed to the side-jump and skew scatter-

ing [21, 23] mechanisms). However, for the present alloy series which behave almost as gapless semiconductors within the whole topologically non-trivial regime, the extrinsic contributions must be small, since the DOS at the Fermi energy, which supplies the electrons for the corresponding scattering is very small as well. As it follows from Fig. 2 (a), within topologically non-trivial regime the value of σ_{xy}^\uparrow reasonably agrees with the quantum of the spin-Hall conductivity in 3D [24, 25] (marked by a blue line in Fig. 2 (a)). The latter is calculated as $\frac{1}{2} \frac{e^2}{2\pi h} K$, where $K = \pi/a$ is the size of the magnetic Brillouin zone in z direction. The factor $1/2$ accounts for the single spin-channel. Since the lattice constant a decreases as a function of x , the quantum of the conductivity slightly increases with rising Hg concentration until the topological phase transition.

IV. CONCLUSIONS

In this paper we have shown that the topological phase transition induced via disorder can be efficiently studied in bulk 3D materials, without the need to investigate large systems, circumventing computationally quite heavy surface calculations. The main conclusion of our study is based on the composition dependence of the spin-Hall ratio $\sigma_{xy}^\uparrow / \sigma_{xx}$ shown in Fig. 2 (b): the ratio nearly scales linearly up with increasing disorder in the system. This behavior can be exploited as an efficient tool for adjusting the adiabatic transport characteristics. The upper limit of such an adjustment can obviously be achieved in the case of full Anderson localization, i. e. when $\sigma_{xx} = 0$. The concept of the Anderson type topological insulator (TAI) joins two very fundamental fields and it is not surprising that it immediately became a subject of intensive theoretical research. The possibility of the TAI state was indicated by C. Beenakker: *Anderson insulator exists due to disorder, whereas topological insulator exists in spite of disorder*. On a model level, provided that the Anderson localization exists, the possibility for the TAI state has already been justified [26–28]. However, even though the Anderson model of localization is rather simple [29, 30], the question whether the full localization can be adequately described in 3D solids within CPA-like theories remains the subject of vivid debates [31–37].

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